

Field induced antiferromagnetism and ^{17}O Knight shift anomaly in La_2CuO_4

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We address the effect of the field induced antiferromagnetism in paramagnetic state of the cuprate weak ferromagnet La_2CuO_4 . The planar oxygen ^{17}O Knight shift is shown to be an effective tool to inspect the effects of Dzyaloshinsky-Moriya coupling in cuprates in an external magnetic field. Field induced antiferromagnetism and anisotropic antiferromagnetic contribution to ^{17}K explain the anomalies observed in ^{17}O NMR in La_2CuO_4 . The experimental observation of antiferromagnetic contribution to the ^{17}O Knight shift provides probably the only way to find out the problem of the sense of Dzyaloshinsky vector in cuprates.

Starting from pioneer papers by Dzyaloshinsky¹ and Moriya² the Dzyaloshinsky-Moriya (DM) antisymmetric exchange coupling was extensively investigated in 60-80ths in connection with weak ferromagnetism focusing on hematite $\alpha\text{-Fe}_2\text{O}_3$ and orthoferrites RFeO_3 .³ The stimulus to a renewed interest to the subject was given by the cuprate problem, in particular, by the weak ferromagnetism observed in La_2CuO_4 ⁴ and many other interesting effects for the DM systems, in particular, the "field-induced gap" phenomena⁵ and field-induced staggered spin polarization.⁶ It is worth noting that the latter effect was addressed earlier on in weak ferromagnet FeF_3 .⁷

Below, in the paper we address the effect of the field-induced antiferromagnetism in La_2CuO_4 and its manifestation in the ^{17}O NMR. We show that namely this effect explains the puzzlingly large negative ^{17}O Knight shift for planar oxygens with anisotropy resembled that of weak ferromagnetism.⁸

At variance with typical 3D systems such as orthoferrites or fluorides, cuprates are characterised by a low-dimensionality, large diversity of Cu-O-Cu bonds including corner- and edge-sharing, different ladder configurations, strong quantum effects for $s = 1/2$ Cu^{2+} centers, and a particularly strong Cu-O covalency resulting in a comparable magnitude of hole charge/spin densities on copper and oxygen sites.

We start with a typical for cuprates the three-center

($\text{Cu}_1\text{-O-Cu}_2$) two-hole system with tetragonal Cu on-site symmetry and ground Cu $3d_{x^2-y^2}$ states (see Fig. 1) which conventional bilinear spin Hamiltonian is written in terms of copper spins as follows

$$\hat{H}_s(12) = J_{12}(\hat{\mathbf{s}}_1 \cdot \hat{\mathbf{s}}_2) + \mathbf{D}_{12} \cdot [\hat{\mathbf{s}}_1 \times \hat{\mathbf{s}}_2] + \hat{\mathbf{s}}_1 \vec{\mathbf{K}}_{12} \hat{\mathbf{s}}_2, \quad (1)$$

where $J_{12} > 0$ is an exchange integral, \mathbf{D}_{12} is the Dzyaloshinsky vector, $\vec{\mathbf{K}}_{12}$ is a symmetric second-rank tensor of the anisotropy constants. In contrast with $J_{12}, \vec{\mathbf{K}}_{12}$, the Dzyaloshinsky vector \mathbf{D}_{12} is antisymmetric with regard to the site permutation: $\mathbf{D}_{12} = -\mathbf{D}_{21}$. Usually this vector is assumed to be located on the bond connecting spins 1 and 2, though, strictly speaking, this should be written as a sum of three vectors located on Cu_1 , Cu_2 , and oxygen sites, respectively: $\mathbf{D}_{12} = \mathbf{D}_{12}^{(1)} + \mathbf{D}_{12}^{(O)} + \mathbf{D}_{12}^{(2)}$. Hereafter we will denote $J_{12} = J, \vec{\mathbf{K}}_{12} = \vec{\mathbf{K}}, \mathbf{D}_{12} = \mathbf{D}$, respectively. It should be noted that making use of effective spin Hamiltonian (1) implies a removal of orbital degree of freedom that calls for a caution with DM coupling as it changes both a spin multiplicity, and an orbital state.

For a composite two $s = 1/2$ spins system one should consider three types of the vector order parameters:

$$\hat{\mathbf{S}} = \hat{\mathbf{s}}_1 + \hat{\mathbf{s}}_2; \quad \hat{\mathbf{V}} = \hat{\mathbf{s}}_1 - \hat{\mathbf{s}}_2; \quad \hat{\mathbf{T}} = 2[\hat{\mathbf{s}}_1 \times \hat{\mathbf{s}}_2] \quad (2)$$

with a kinematic constraint:

$$\hat{\mathbf{S}}^2 + \hat{\mathbf{V}}^2 = 3\hat{\mathbf{I}}; \quad (\hat{\mathbf{S}} \cdot \hat{\mathbf{V}}) = 0; \quad (\hat{\mathbf{T}} \cdot \hat{\mathbf{V}}) = 6i; \quad [\hat{\mathbf{T}} \times \hat{\mathbf{V}}] = \hat{\mathbf{S}}. \quad (3)$$

In a sense the $\hat{\mathbf{V}}$ operator describes the effect of local antiferromagnetic order, while $\hat{\mathbf{T}}$ operator may be associated with a vector chirality.⁹ In recent years, phases with broken vector chirality in frustrated quantum spin chains have attracted considerable interest. Such phases are characterized by nonzero long-range correlations of the vector order parameter $\langle \hat{\mathbf{T}} \rangle$. Interestingly that a chirally ordered phase can manifest itself as a "nonmagnetic" one, with $\langle \hat{\mathbf{S}} \rangle = \langle \hat{\mathbf{V}} \rangle = 0$.

Both $\hat{\mathbf{T}}$ and $\hat{\mathbf{V}}$ operators change the spin multiplicity with matrix elements

$$\langle 00 | \hat{T}_m | 1n \rangle = -\langle 1n | \hat{T}_m | 00 \rangle = i\delta_{mn};$$

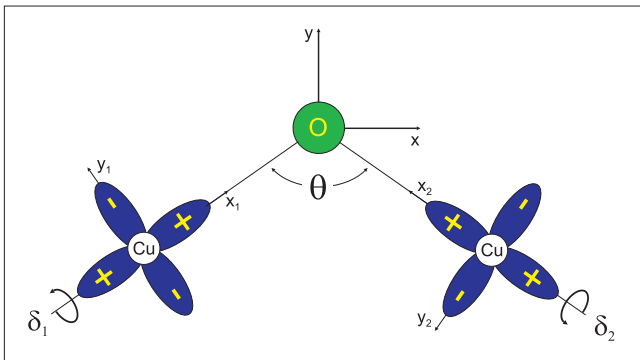


FIG. 1: Geometry of the three-center (Cu-O-Cu) two-hole system with ground Cu $3d_{x^2-y^2}$ states.

$$\langle 00|\hat{V}_m|1n\rangle = \langle 1n|\hat{V}_m|00\rangle = \delta_{mn}, \quad (4)$$

where we made use of Cartesian basis for $S = 1$. The eigenstates of the operators \hat{V}_n , and \hat{T}_n with nonzero eigenvalues ± 1 form Néel doublets $\frac{1}{\sqrt{2}}(|00\rangle \pm |1n\rangle)$ and DM doublets $\frac{1}{\sqrt{2}}(|00\rangle \pm i|1n\rangle)$, respectively. The Néel doublets correspond to classical collinear antiferromagnetic spin configurations, while the DM doublets correspond to quantum spin configurations which sometimes are associated with a rectangular 90° spin ordering in the plane orthogonal to the Dzyaloshinsky vector.

Before going to microscopic analysis we should note that the interaction of our three-center system with external spins and/or fields \hat{H}_{ext} is usually addressed by introducing only two types of effective external fields: the conventional Zeeman-like field and unconventional Néel-like staggered field, so that \hat{H}_{ext} reads as follows

$$\hat{H}_{ext} = -(\mathbf{h}^S \cdot \hat{\mathbf{S}}) - (\mathbf{h}^V \cdot \hat{\mathbf{V}}). \quad (5)$$

It should be noted that an ideal Néel state is attainable only in the limit of infinitely large staggered field, therefore for a finite staggered field $\mathbf{h}^V \parallel \mathbf{n}$ the ground state is a superposition of a spin singlet and a Néel state,

$$\Psi = \cos \alpha |00\rangle + \sin \alpha |1n\rangle, \quad \tan 2\alpha = \frac{2h^V}{J},$$

which composition reflects the role of quantum effects. For instance, in a Heisenberg spin $1/2$ chain with nn exchange the maximal value of staggered field $h^V = J/2$ hence the Ψ function strongly differs from that of Néel state ($\langle \hat{V}_n \rangle = \sin 2\alpha = \frac{1}{\sqrt{2}}$), and quantum mechanical average for a single spin $\langle s_z \rangle \leq \frac{1}{2} \sin \pi/4 = \frac{1}{\sqrt{2}} \cdot \frac{1}{2} \approx 0.71 \cdot \frac{1}{2}$ deviates strongly from classical value $\frac{1}{2}$. It should be noted that for the isolated antiferromagnetically coupled spin pair the zero-temperature uniform spin susceptibility turns into zero: $\chi^S = 0$, while for the staggered spin susceptibility we obtain $\chi^V = 2/J$.

Application of an uniform external magnetic field \mathbf{h}_S will produce a staggered spin polarization in the antiferromagnetically coupled $\text{Cu}_1\text{-Cu}_2$ pair

$$\langle \mathbf{V}_{12} \rangle = \mathbf{L} = -\frac{1}{J_{12}^2} \left[\sum_i \mathbf{D}_{12}^{(i)} \times \mathbf{h}^S \right] = \overset{\leftarrow}{\chi}^{VS} \mathbf{h}^S \quad (6)$$

with antisymmetric VS -susceptibility tensor: $\chi_{\alpha\beta}^{VS} = -\chi_{\beta\alpha}^{VS}$. One sees that the sense of a staggered spin polarization, or antiferromagnetic vector, depends on that of Dzyaloshinsky vector.⁷ The VS coupling results in many interesting effects for the DM systems, in particular, the "field-induced gap" phenomena in 1D $s=1/2$ antiferromagnetic Heisenberg system with alternating DM coupling.⁵ Approximately, the phenomenon is described by a so called *staggered* $s=1/2$ antiferromagnetic Heisenberg model with the Hamiltonian

$$\hat{H} = J \sum_i (\hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+1}) - h_u \hat{S}_{iz} - (-1)^i h_s \hat{S}_{ix}, \quad (7)$$

which includes the effective uniform field h_u and the induced staggered field $h_s \propto h_u$ perpendicular both to the applied uniform magnetic field and Dzyaloshinsky vector.

Earlier on (see Ref.7) we pointed to the ligand NMR as, probably, the only experimental technique to measure both staggered spin polarization, or antiferromagnetic vector in weak 3d-ferromagnets and the value, direction, and the sense of Dzyaloshinsky vector. The latter possibility was realized with ^{19}F NMR for weak ferromagnet FeF_3 .⁷ Here we address the problem for generic cuprate weak ferromagnet La_2CuO_4 . The DM coupling and magnetic anisotropy in La_2CuO_4 and related compounds has attracted considerable attention in 90-ths (see, e.g., Refs.10,11,12,13), and is still debated in the literature.^{14,15} In the low-temperature tetragonal (LTT) and orthorhombic (LTO) phases of La_2CuO_4 , the oxygen octahedra surrounding each copper ion rotate by a small tilting angle ($\delta_{LTT} \approx 3^\circ$, $\delta_{LTO} \approx 5^\circ$) relative to their location in the high-temperature tetragonal (HTT) phase. The structural distortion allows for the appearance of the antisymmetric Dzyaloshinsky-Moriya interaction. In terms of our choice for structural parameters to describe the $\text{Cu}_1\text{-O-Cu}_2$ bond we have for LTT phase: $\theta = \pi$; $\delta_1 = \delta_2 = \frac{\pi}{2} \pm \delta_{LTT}$ for bonds oriented perpendicular to the tilting plane, and $\theta = \pm(\pi - 2\delta_{LTT})$; $\delta_1 = \delta_2 = \frac{\pi}{2}$ for bonds oriented parallel to the tilting plane. It means that all the local Dzyaloshinsky vectors turn into zero for the former bonds, and turn out to be perpendicular to the tilting plane for the latter bonds. For LTO phase: $\theta = \pm(\pi - \sqrt{2}\delta_{LTO})$; $\delta_1 = \delta_2 = \frac{\pi}{2} \pm \delta_{LTO}$. The largest ($\propto \delta_{LTO}$) component of the local Dzyaloshinsky vectors (z-component in our notation) turns out to be oriented perpendicular to the $\text{Cu}_1\text{-O-Cu}_2$ bond plane. Other two components of the local Dzyaloshinsky vectors are fairly small: that of perpendicular to CuO_2 plane (y-component in our notation) is of the order of δ_{LTO}^2 , while that of oriented along $\text{Cu}_1\text{-Cu}_2$ bond axis (x-components in our notation) is of the order of δ_{LTO}^3 .

As an important by-product of the cuprate activity we arrived at a significant progress in different experimental methods and new opportunities to elucidate subtle details of electron and spin structure. In particular, it concerns the oxygen ^{17}O NMR-NQR as an unique local probe to study the charge and spin densities on oxygen sites. In this connection we point to papers by R. Walstedt *et al.*⁸ as a first direct observation of anomalous oxygen hyperfine interactions in generic cuprate La_2CuO_4 . With the approaching transition to the ordered magnetic phase, the authors observed anomalously large negative ^{17}O Knight shift for planar oxygens which anisotropy resembled that of weak ferromagnetism in this cuprate. The giant shift was observed only when external field was parallel to the local Cu-O-Cu bond axis (PL1 lines) or perpendicular to CuO_2 plane. The effect was not observed for PL2 lines which correspond to oxygens in the local Cu-O-Cu bonds which axis is perpendicular to in-plane external field. The data were interpreted as an indication of oxygen spin polarization due to a local

Dzyaloshinsky-Moriya antisymmetric exchange coupling. However, either interpretation of NMR-NQR data in such low-symmetry systems as La_2CuO_4 needs in a thorough analysis of transferred hyperfine interactions and a revisit of some textbook results being typical for the model high-symmetry systems. First we draw attention to spin-dipole hyperfine interactions for O 2p-holes which are main participants of $\text{Cu}_1\text{-O-Cu}_2$ bonding. Starting from a conventional formula for a spin-dipole contribution to local field

$$\mathcal{H}_n = -g_s\mu_B \sum_i \frac{3(\mathbf{r}_i \cdot \mathbf{s}_i)\mathbf{r}_i - r_i^2\mathbf{s}_i}{r_i^5}$$

and making use of an expression for appropriate matrix element

$$\begin{aligned} \langle p_i | \frac{3x_\alpha x_\beta - r^2 \delta_{\alpha\beta}}{r^5} | p_j \rangle &= -\frac{2}{5} \left\langle \frac{1}{r^3} \right\rangle_{2p} \langle p_i | 3\widetilde{l_\alpha l_\beta} - 2\delta_{\alpha\beta} | p_j \rangle \\ &= \frac{2}{5} \left\langle \frac{1}{r^3} \right\rangle_{2p} \left(\frac{3}{2} \delta_{\alpha i} \delta_{\beta j} + \frac{3}{2} \delta_{\alpha j} \delta_{\beta i} - \delta_{\alpha\beta} \delta_{ij} \right) \end{aligned} \quad (8)$$

we present a local field on the ^{17}O nucleus in $\text{Cu}_1\text{-O-Cu}_2$ system as a sum of ferro- and antiferromagnetic contributions as follows⁷

$$\mathcal{H}_n = \overset{\leftrightarrow S}{\mathbf{A}} \cdot \langle \hat{\mathbf{S}} \rangle + \overset{\leftrightarrow V}{\mathbf{A}} \cdot \langle \hat{\mathbf{V}} \rangle \quad (9)$$

where

$$\overset{\leftrightarrow S}{\mathbf{A}} = \overset{\leftrightarrow S}{\mathbf{A}}(dp) + \overset{\leftrightarrow S}{\mathbf{A}}(pd); \quad \overset{\leftrightarrow V}{\mathbf{A}} = \overset{\leftrightarrow V}{\mathbf{A}}(pd) - \overset{\leftrightarrow V}{\mathbf{A}}(dp),$$

$$A_{ij}^S(dp) = A_p^{(0)} [3c_t(dp_i)c_t(dp_j) - |\mathbf{c}_t(dp)|^2 \delta_{ij}],$$

$$A_{ij}^S(pd) = A_p^{(0)} [3c_t(p_i d)c_t(p_j d) - |\mathbf{c}_t(pd)|^2 \delta_{ij}],$$

$$A_{ij}^V(dp) = A_p^{(0)} [3c_s(dp_i)\widetilde{c_t(dp_j)} - (\mathbf{c}_s(dp) \cdot \mathbf{c}_t(dp))\delta_{ij}],$$

$$A_{ij}^V(pd) = A_p^{(0)} [3c_s(p_i d)\widetilde{c_t(p_j d)} - (\mathbf{c}_s(pd) \cdot \mathbf{c}_t(pd))\delta_{ij}],$$

where

$$\begin{aligned} c_{s,t}(dp_x) &= -\frac{\sqrt{3}}{2} \frac{t_{dp\sigma}}{E_{s,t}(dp_x)} \sin \frac{\theta}{2}, \\ c_{s,t}(dp_y) &= -\frac{\sqrt{3}}{2} \frac{t_{dp\sigma}}{E_{s,t}(dp_y)} \cos \frac{\theta}{2} \end{aligned} \quad (10)$$

are probability amplitudes for different singlet (c_s) and triplet (c_t) 110 ($\text{Cu}_1 3d_{x^2-y^2} \text{O} 2p_{x,y}$) and 011 ($\text{O} 2p_{x,y} \text{Cu}_2 3d_{x^2-y^2}$) configurations in the ground state wave function, respectively; $c_{s,t}(dp_x) = -c_{s,t}(p_x d)$,

$c_{s,t}(dp_y) = c_{s,t}(p_y d)$, $t_{dp\sigma}$ is a hole dp-transfer integral, $A_p^{(0)} = \frac{2}{5} g_s \mu_B \langle \frac{1}{r^3} \rangle_{2p}$, the tilde points to a symmetrization. The energies $E_{s,t}(dp_{x,y})$ are those for singlet and triplet states of $dp_{x,y}$ configurations, respectively: $E_{s,t}(dp_{x,y}) = \epsilon_{x,y} + K_{dp_{x,y}} \pm I_{dp_{x,y}}$, where $K_{dp_{x,y}}$ and $I_{dp_{x,y}}$ are Coulomb and exchange dp-integrals, respectively. Thus, along with a conventional textbook ferromagnetic ($\propto \langle \hat{\mathbf{S}} \rangle$) transferred hyperfine contribution to local field which simply mirrors a sum total of two Cu-O bonds, we arrive at an additional unconventional antiferromagnetic difference ($\propto \langle \hat{\mathbf{V}} \rangle$) contribution which symmetry and magnitude strongly depend on the orientation of the oxygen crystal field axes and $\text{Cu}_1\text{-O-Cu}_2$ bonding angle. In the case of $\text{Cu}_1\text{-O-Cu}_2$ geometry shown in Fig.1 we arrive at a diagonal $\overset{\leftrightarrow S}{\mathbf{A}}$ tensor:

$$\begin{aligned} A_{xx}^S &= 2A_p(3\sin^2 \frac{\theta}{2} - 1); \quad A_{yy}^S = 2A_p(3\cos^2 \frac{\theta}{2} - 1); \\ A_{zz}^S &= -2A_p, \end{aligned} \quad (11)$$

and the only nonzero components of $\overset{\leftrightarrow V}{\mathbf{A}}$ tensor:

$$A_{xy}^V = A_{yx}^V = 3A_p \sin \theta \quad (12)$$

with

$$A_p = \frac{3}{4} \left(\frac{t_{dp\sigma}}{\epsilon_p} \right)^2 A_p^0 = f_\sigma A_p^0, \quad (13)$$

where f_σ is the parameter of a transferred spin density and we made use of a simple approximation $E_{s,t}(dp_{x,y}) \approx \epsilon_p$. Generally speaking, we should take into account an additional contribution of magneto-dipole hyperfine interactions.

The two-term structure of oxygen local field implies a two-term S-V structure of the ^{17}O Knight shift

$$^{17}\text{K} = \overset{\leftrightarrow S \leftrightarrow SS}{\mathbf{A}} \chi + \overset{\leftrightarrow V \leftrightarrow VS}{\mathbf{A}} \chi \quad (14)$$

that points to Knight shift as an effective tool to inspect both uniform and staggered spin polarization. The existence of antiferromagnetic term in oxygen hyperfine interactions yields a rather simple explanation of the ^{17}O Knight shift anomalies in La_2CuO_4 ⁸ as a result of the external field induced staggered spin polarization $\langle \hat{\mathbf{V}} \rangle = \mathbf{L} = \overset{\leftrightarrow VS}{\chi} \mathcal{H}_{\text{ext}}$. Indeed, "our" local y axis for $\text{Cu}_1\text{-O-Cu}_2$ bond corresponds to the crystal tetragonal c -axis oriented perpendicular to CuO_2 planes both in LTO and LTT phases of La_2CuO_4 while x -axis does to local Cu-O-Cu bond axis. It means that for the geometry of the experiment by Walstedt *et al.*⁸ (the crystal is oriented so that the external uniform field is either \parallel or \perp to the local Cu-O-Cu bond axis) the antiferromagnetic contribution to ^{17}O Knight shift will be observed only a) for oxygens in $\text{Cu}_1\text{-O-Cu}_2$ bonds oriented along

external field or b) for external field along tetragonal c -axis. Experimental data⁸ agree with staggered magnetization along the tetragonal c -axis in the former and along the rhombic c -axis (tetragonal ab -axis) in the latter. Interestingly, the sizeable effect has been observed in La_2CuO_4 for temperatures $T \sim 500$ K that is essentially higher than $T_N \approx 300$ K. Given $L = 1$, $A_p^{(0)} \approx 100$ kG/spin,⁸ $|\sin \theta| \approx 0.1$, and $f_\sigma \approx 20\%$ we obtain ≈ 6 kG as a maximal value of a low-temperature antiferromagnetic contribution to hyperfine field which is parallel to external magnetic field. This value agrees with a low-temperature extrapolation of high-temperature experimental data by Walstedt *et al.*⁸ Similar effect of anomalous ^{13}C Knight shift has recently been observed in copper pyrimidine dinitrate $[\text{CuPM}(\text{NO}_3)_2(\text{H}_2\text{O})_2]_n$, a one-dimensional $S=1/2$ antiferromagnet with alternating local symmetry.⁶ However, the authors did take into account only the inter-site magneto-dipole contribution to \vec{A}^V tensor that questions their quantitative conclusions regarding the "giant" spin canting.

The ferro-antiferromagnetic S-V structure of local field on the nucleus of an intermediate oxygen ion in a $\text{Cu}_1\text{-O-Cu}_2$ triad points to ^{17}O NMR as, probably, the only experimental technique to measure both the value, direction, and the sense of Dzyaloshinsky vector. For instance, the negative sign of ^{17}O Knight shift in La_2CuO_4 ⁸ points

to a negative sign of $\chi^{\leftrightarrow VS}$ for $\text{Cu}_1\text{-O-Cu}_2$ triad with $A_{xy}^V > 0$, hence to a positive sense of z -component of the summary Dzyaloshinsky vector in $\text{Cu}_1\text{-O-Cu}_2$ triad with geometry shown in Fig.1 given $\theta \leq \pi$, $\delta_1 = \delta_2 \approx \pi/2$. It should be emphasized that the above effect is determined by the summary Dzyaloshinsky vector in $\text{Cu}_1\text{-O-Cu}_2$ triad rather than by a local oxygen "weak-ferromagnetic" polarization as it was proposed by Walstedt *et al.*⁸

In conclusion, we predict the effect of the field induced antiferromagnetism in a paramagnetic state of the cuprate weak ferromagnet La_2CuO_4 . The planar oxygen ^{17}O Knight shift is shown to be an effective tool to inspect the effects of Dzyaloshinsky-Moriya coupling in cuprates in an external magnetic field. Field induced antiferromagnetism and anisotropic antiferromagnetic contribution to ^{17}K explain the anomalies observed in ^{17}O NMR in La_2CuO_4 .⁸ The experimental observation of antiferromagnetic contribution to the ^{17}O Knight shift provides probably the only way to find out the problem of the sense of Dzyaloshinsky vector in cuprates.

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